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Materials Impurity Analysis by Means of Nuclear Resonance Reactions

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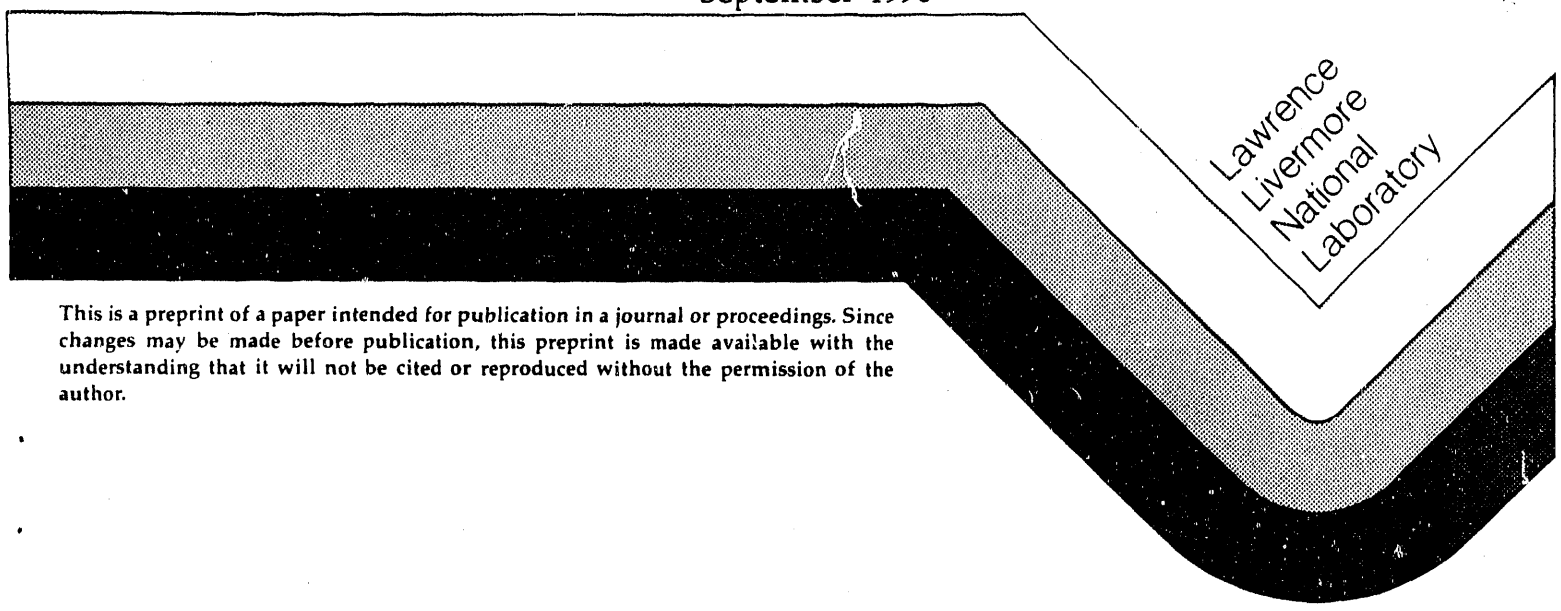

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Abstract

An automated beam line for measurement of trace impurities in materials is installed on the FN tandem in the Center for Accelerator Mass Spectrometry, LLNL. The major purpose of the beam-line system is to determine the diffusion of water into glasses by use of the $^{19}F(p,\alpha\gamma)^{16}O$ reaction. This study is stimulated by the radiation-waste-isolation program; however, the system is applicable to studies of other impurities in many material matrices. The target wheels, each of which holds twenty-four samples, can be changed in less than an hour. The beam-line control system both indexes the target wheel and controls the tandem energy as it steps through the requested measurement protocol. The tandem control system, in response to a request, adjusts the terminal potential and other transport elements to maintain constant spot size and location.

I. Introduction

The proposal to encapsulate high-level radioactive waste in glasses led to the search for the optimal glass for the purpose. Studies of expected storage environments suggested that resistance to dissolution in hot brine is a criterion. In order to measure the rate of dissolution by water of candidate glasses, this beam line was constructed. Water apparently attacks glass by losing a hydrogen atom which migrates into the glass, upsetting silicate bonds as it diffuses. A measurement of the hydrogen density versus depth allows the calculation of the diffusion coefficient, from which the lifetime of the glass in service is estimated.

The hydrogen density is measured, in this case, by the threshold resonance reaction with fluorine [$^{19}F(p,\alpha\gamma)^{16}O$]. The threshold is at 16.44 MeV and the resonance has a width of about 40 keV. The gamma rays have energy of 6.13, 6.92 and 7.12 MeV. The gamma rays are easy to detect and the sharp resonance leads to good analytical sensitivity. Nitrogen is also commonly used for this purpose, but since radiocarbon dating is a major activity in this facility and that reaction produces ^{14}C, the system could be contaminated.

The need to analyze many samples has led to the design of a facility featuring automatic operation and rapid turnaround. Special attention was given in the design for easy mounting of diverse samples and easy installation of a target wheel. The samples are

large with respect to the beam spot size (a few hundred microns) and homogeneous. Because there is no interest in making local measurements across a single sample, the target indexing registration is not carefully controlled. The softwear has user features to support processing of large numbers of samples. For example, a file of data which describes the targets in a sample wheel can be generated offline and saved for future use. Editing facilities make using subsets of the descriptor file straightforward.

II. Hardware Design

Because of the expected target size and the need to process many samples, twenty-four target positions were fit on a twelve-inch diameter wheel. These considerations determine the overall scale of the chamber. For optimum depth resolution in the sample, the energy resolution must be excellent. The beam is bent 60 degrees and focused through slits which define the spot size on the target. The slits are placed near the focus point of the deflection magnet and about a meter away from the scintillator detector in order to limit the background. A Faraday cup is installed in the line as a tuning aid for the operator and as a beam stop when targets are being indexed. A beam-line scanner is included to determine the position and shape of the beam just in front of the slits. A six-way cross in front of the chamber provides a pumping port, a window to view the targets, a mount for the vacuum gauges and a mount for electrical feedthroughs. To monitor beam current, the target chamber is isolated and serves as a Faraday cup. To have a large

solid angle for the detector, the back flange of the chamber is re-entrant and the scintillator is located in the well. An integrated scintillator-photomultiplier serves as the detector. The detector mount is insulated from the target chamber. Figure 1 shows some of these features.

To change targets, the chamber is vented and the entire back flange is swung aside. (A single central nut retracts the back flange, a slide latch is released and the chamber is opened.) The target wheel is centered with a cone-shaped nut on a stud on the mounting shaft and a dowel pin provides angular alignment. With this arrangement, a set of targets is changed in essentially the amount of time necessary to pump out the chamber.

To support the stand-alone operation of this system, all parameters and controls are machine readable or both readable and controllable. These controls include the particular target that is in the beam, the in-out position of the Faraday cup, the PMT voltage, and the vacuum level (the pressure is below a predetermined set point). Also all variables are read automatically and accumulated for processing with the data. These include the tandem energy, the ion beam current, and the histogram of the detected gamma rays. The monitor and control features are implemented through CAMAC by means of standard modules. The crate is controlled via a GPIB interface. For communication of the beam energy request to the tandem, pairs of input and output registers are used.

The beam-line control computer writes a tandem voltage request in an output register in the beam-line CAMAC crate. The output register is connected to an input register which is read by the tandem control system. When the tandem control system detects a change of state in the input register, it begins to slew to the new energy value. To prohibit the beam line from having an incorrect energy value, the tandem controller (also equipped with an output register that is connected to an input register in the beam-line system) writes a negative value in the register while it is actively adjusting the beam. When the beam is stabilized, the actual energy value is written by the tandem and read by the beam-line computer. This value is the one used in processing the data. While this may seem a bit clumsy for interfacing, when only one word needs to be communicated, this simple approach avoids issues of computer-to-computer interface and fits seamlessly into the two control systems.

Screen labeled softkeys provide the user interface to the system. An editor accepts input information describing the array of targets and the energy-scan parameters. Input data are checked for validity and completeness as they are entered, and the entire file is also checked before a run is started. The user is prompted by the system if the data are incomplete and also given default values that may be used. The run may be done manually or automatically under computer control. Because the run may be very long if there are many samples, a restart capability is included.

The tandem is not expected to come exactly to the desired energy or repeatedly to the same value, so the energy is held constant while targets are sequentially indexed before the beam. During the indexing, the Faraday cup is inserted and the current scalar is zeroed. Then the cup is removed and a histogram of emitted gamma rays acquired. After a fixed acquisition time, the cup is reinserted and the ADC is gated off. The histogram is transfered to the computer and the current read from the scalar. This sequence is repeated for each target at each energy value. After the arrays are acquired, the data are sorted into a sequence of increasing energy values before being stored.

III. Data analysis

Four distinct experiments are conducted for each hydrogen-profile measurement. These are (1) determination of the background count rate due to environmental factors, (2) determination of the count rate per incident beam particle on a sample with no hydrogen, (3) determination of the count rate with a target of known hydrogen density (to deduce the overall efficiency of the detection system), and (4) measurement of the count rate with the beam on a target containing an unknown amount of hydrogen. The first two measurements estimate the spurious counts for the next two measurements.

The region of interest (ROI) is determined by examining histograms of background counts with and without beam. The ROI

will be used automatically in subsequent processing. After the ROI is established and the environmental count rate determined, the rest of the measurement protocol is done automatically under computer control. To avoid accidental loss of data, after the raw data are acquired they are automatically written to the archive media.

The data are read back from the archive for analysis. The expected number of spurious counts must be subtracted from the raw counts. Next, the efficiency of the detector system per incident beam particle is calculated. Finally, the amount of hydrogen in the unknown sample is deduced for each value of beam energy.

Since the environmental factor is constant in time, a count rate (Nbkg) is actually determined. The sampling time (Ton) is known. The counts of beam on a non-hydrogen target (Nnh) are proportional to the number of beam particles (Nbeam). The corrected counts (Ncc) per incident beam particle are then found from the number of raw counts (Nr) by

$$Ncc=[Nr-(Nbkg*Ton+Nnh*Nbeam)]/Nbeam.$$

The detection efficiency (Eps) is calculated from the known hydrogen density (Hdencal) and cross-section data (Hxsect),

$$Eps=Ncc*Dedx/(Nbeam*Hdencal*Hxsect*Deres).$$

Ultimately, the thickness of the interaction region [i.e., the depth over which the beam is within the the resonance energy band (Deres)] must be determined. The assumption is that the energy-loss rate in the glass (Dedx) is adequately described by the "Trim" code [1], and the thickness is equal to the resonance width divided by the energy-loss rate. At present this is treated as a constant, but the analysis softwear is designed to accommodate any suitable model of the loss rate. The integral of the loss rate also determines the estimate of depth which must be used in calculating the diffusion coefficient. The unknown hydrogen density (Hden) is then reckoned from the above formula using the now known detection efficiency.

Throughout the analysis procedure, error estimates of each process are also calculated and are available to the experimenter.

IV. Conclusion

Initial measurements were made on glass samples. The system has worked as designed. Many impurities in other materials may be analyzed using this technique.

References

[1] J.F. Ziegler, J.P. Biersack, U. Littmark, The Stopping and Range of Ions in Solids, Vol. 1 (Pergamon Press, New York, 1985) 321pp.

Figure Captions

Figure 1. Schematic drawing of the target chamber and associated hardwear.

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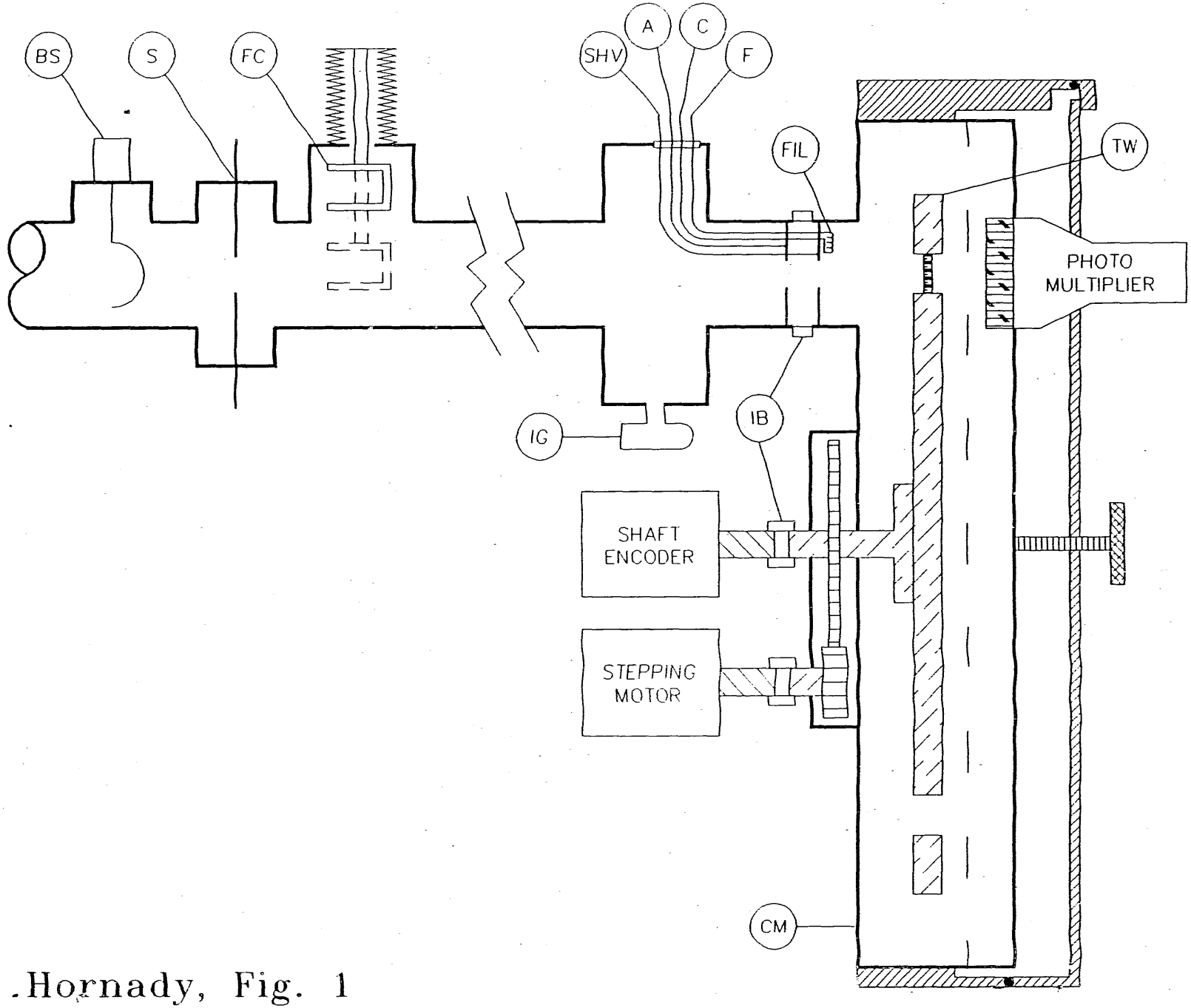

R. S. Hornady, Fig. 1

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